

Predictors of Indoor Radon Concentrations in Pennsylvania, 1989–2013

Joan A. Casey, Elizabeth L. Ogburn, Sara G. Rasmussen,
Jennifer K. Irving, Jonathan Pollak, Paul A. Locke,
and Brian S. Schwartz

<http://dx.doi.org/10.1289/ehp.1409014>

Received: 30 July 2014

Accepted: 31 March 2015

Advance Publication: 9 April 2015

Note: When this article was originally posted, the word “declined” was inadvertently omitted from the statement “When we included multiple basement measurements per building, radon levels [declined] with repeated measurements within a building, which is good news for public health and suggests state remediation programs are effective.” That typographic error has been corrected in this PDF.

This article will be available in its final, 508-conformant form 2–4 months after Advance Publication. If you need assistance accessing this article before then, please contact ehp508@niehs.nih.gov. Our staff will work with you to assess and meet your accessibility needs within 3 working days.

Predictors of Indoor Radon Concentrations in Pennsylvania, 1989–2013

Joan A. Casey,¹ Elizabeth L. Ogburn,² Sara G. Rasmussen,¹ Jennifer K. Irving,³ Jonathan Pollak,¹ Paul A. Locke,^{1,4} and Brian S. Schwartz^{1,3,5}

¹Department of Environmental Health Sciences, Johns Hopkins Bloomberg School of Public Health, Baltimore, Maryland, USA; ²Department of Biostatistics, Johns Hopkins Bloomberg School of Public Health, Baltimore, Maryland, USA; ³Center for Health Research, Geisinger Health System, Danville, Pennsylvania, USA; ⁴Department of Health Policy and Management, Johns Hopkins Bloomberg School of Public Health, Baltimore, Maryland, USA; ⁵Department of Medicine, Johns Hopkins School of Medicine, Baltimore, Maryland, USA

Address correspondence to Brian S. Schwartz, Johns Hopkins Bloomberg School of Public Health, 615 N. Wolfe Street, Room W7041, Baltimore, MD 21205 USA. E-mail:

bschwar1@jhu.edu

Running title: Predictors of indoor radon concentrations

Acknowledgments: R.K. Lewis at the Pennsylvania Department of Environmental Protection facilitated acquisition of the radon data. J.M. Crisp assisted with the assembling and cleaning of the Marcellus well data. R.D. Peng provided statistical support.

Research supported in part by the National Institutes of Health R21 ES023675.

Competing financial interests: The authors declare that they have no actual or potential competing financial interests.

Abstract

Background: Radon is the second-leading cause of lung cancer worldwide. Most indoor exposure occurs by diffusion of soil gas. Radon is also found in well water, natural gas and ambient air. Pennsylvania has high indoor radon concentrations; buildings are often tested during real estate transactions with results reported to the Department of Environmental Protection (PADEP).

Objectives: To evaluate predictors of indoor radon concentrations.

Methods: Using first floor and basement indoor radon results reported to the PADEP between 1987-2013, we evaluated associations of radon concentrations (*ln*-transformed) with geology, water source, building characteristics, season, weather, community socioeconomic status, community type and unconventional natural gas development measures based on drilled and producing wells.

Results: Primary analysis included 866,735 first measurements by building, the large majority from homes. The geologic rock layer on which the building sat was strongly associated with radon concentration (e.g., Axemann Formation, median = 365 Bq/m³, IQR = 167-679 vs. Stockton Formation, median = 93 Bq/m³, IQR = 52-178). In adjusted analysis, buildings using well water had 21% higher concentrations ($\beta = 0.191$, 95% CI: 0.184, 0.198). Buildings in cities (vs. townships) had lower concentrations ($\beta = -0.323$, 95% CI: -0.333, -0.314). When we included multiple tests per building, concentrations declined with repeated measurements over time. Between 2005-2013, 7469 unconventional wells were drilled in Pennsylvania. Basement radon concentrations fluctuated between 1987-2003, but began an upward trend from 2004-2012 in all county categories ($p < 0.001$), higher levels in counties with ≥ 100 drilled wells vs. counties with none, and with highest levels in the Reading Prong.

Conclusions: Geologic unit, well water, community, weather and unconventional natural gas development were associated with indoor radon concentrations. Future studies should include direct environmental measurement of radon, and building features unavailable for this analysis.

Introduction

Exposure to radon-222 – an inert, odorless, and carcinogenic gas – is the second leading cause of lung cancer worldwide (Darby et al. 2005; Pawel and Puskin 2004). The Environmental Protection Agency (EPA) estimates that indoor radon exposure causes or contributes to about 21,000 lung cancer deaths in the U.S. annually (Pawel and Puskin 2004). In 1986, the EPA set an action level of 148 Bq/m³ (4 pCi/L; there are 37 Bq/m³ per pCi/L) based on the current state of radon testing and mitigation technologies (EPA 1992; NAS 1999).

Uranium-238 occurs naturally in soil and bedrock and decays to radium-226, which decays to radon. Both uranium-238 and radium-226 persist in the environment (half-lives 4.5 billion years and 1,600 years, respectively). Radon-222 has a half-life of 3.8 days and its radioactive decay products are responsible for its carcinogenicity. Pressure differentials between soil gas and indoor air cause the migration of radon through cracks and other openings into buildings, the primary source of indoor radon. Radium and radon are soluble in water, with concentrations increasing as salinity increases (Warner et al. 2012).

Several counties in eastern Pennsylvania overlie the Reading Prong, a physiographic section known to have high bedrock uranium concentrations (Gundersen 1991) and elevated indoor radon levels. The entire state has had some of the highest indoor radon levels in the U.S. The Pennsylvania Department of Environmental Protection (PADEP) established a Radon Division that administers a program of radon monitoring and remediation (http://www.portal.state.pa.us/portal/server.pt/community/radon_division/21923).

U.S. Geological Survey (USGS) analysis of 548,547 indoor and short-term radon test results compiled by the PADEP from 1990-2007 reported that 39% of radon tests exceeded the EPA

action level and that concentrations varied dramatically by geologic unit, a rock layer of a given lithology and geologic period (e.g., Annville Formation, high-calcium limestone from the Ordovician period) (Gross 2013). Geologists have identified 195 geologic units in Pennsylvania. Other factors that have been associated with higher indoor radon levels include the use of radon-rich well water (Folger et al. 1994; UNSCEAR 2009), colder months, less precipitation, more expensive housing, rural area, and higher individual socioeconomic status (SES) (Cohen and Gromicko 1988; Folger et al. 1994; UNSCEAR 2009). Radon is present in natural gas used for cooking and heating; calculations performed in the 1970s suggested that it would not be expected to result in an increase in indoor radon levels (Johnson Jr et al. 1973). Radon can also enter buildings from ambient air, however outdoor radon concentrations are generally low, around 10 Bq/m³, but can range from 1-100 Bq/m³ (UNSCEAR 2009).

The development of unconventional natural gas in the Marcellus shale in Pennsylvania has the potential to exacerbate several pathways for entry of radon into buildings. The USGS reported 91,020 Bq/m³ (n = 14) as the median radium concentration in produced water from Marcellus wells (Rowan et al. 2011), a value nearly 500 times the federal drinking water limit (185 Bq/m³) and one that far exceeds the industrial discharge limit of 2220 Bq/m³. Underground, radon collects in porous geological formations and thus in natural gas production wells (Gogolak 1980). Shales also tend to contain both higher concentrations of uranium (3.7-40ppm) than other geologic formations and higher concentrations of radon in their natural gas (Gogolak 1980). The USGS reported preliminary data from 11 wellheads in Pennsylvania with corrected concentrations of radon (devices were calibrated for air measurement, but used in natural gas with correction factor = gas measurement x 1.47) ranging from 37-2923 Bq/m³ (median = 1369)

(Rowan and Kraemer 2012), suggesting that shale gas may have higher radon levels than other natural gas sources.

To our knowledge, no prior studies have evaluated predictors of radon concentrations in Pennsylvania. Our main objective was to identify the independent contribution to indoor radon concentrations of geologic unit, water source, building characteristics, season, weather, community SES, community type and Marcellus shale development.

Methods

Study design

We obtained radon data on 1,983,705 indoor radon tests conducted in the state in 806,469 buildings between 1987-2013 from all 67 counties that were submitted by certified testers, laboratories, or homeowners to the PADEP Bureau of Radiation Protection, Radon Division. Buildings are most often tested during real estate transactions (WHO 2009) and the PADEP requires reporting of all test results to their GreenPort website (<http://www.depgreenport.state.pa.us/>). We used the subset of radon measurements taken between January 1, 1989 and December 31, 2013 in our analysis because few samples were available from earlier years (n = 4294) (Figure 1). The Institutional Review Board at the Johns Hopkins Bloomberg School of Public Health reviewed the study protocol and did not consider it to be human subjects research.

Outcome: indoor radon concentration

Data included the address of the tested building, building type (12 types, Table 1), test location (i.e. basement, first floor, second floor), test type (i.e. activated charcoal, alpha-track detectors, charcoal liquid scintillation, continuous radon monitors, electret ion chamber), test dates, and

radon concentration (Bq/m^3). Results were available for both short-term (2-7 days) and long-term (up to one year) testing periods. We used ArcGIS (version 10.0, Esri) and 10 street maps (e.g. TeleAtlas, TIGER files, and StreetMap Premium from 2000-2012) to obtain latitude and longitude of buildings.

We excluded tests from buildings that could not be geocoded to an address, that were out of state, that were not taken on the first floor or basement, or that appeared in the database more than once ($n = 394,008$ buildings). Many buildings ($n = 307,245$) had multiple radon measurements (range 2-56) taken on the same floor and day. For example, in buildings with 2 measurements per floor ($n = 291,098$) the correlation of floor-specific measurements was very high ($\rho = 0.91$). Because we had no information on building remediation, our primary analysis included only measurements taken during the first test day at each building ($n = 866,735$, including $n = 224,666$ averaged concentrations from same floor and day). In a sensitivity analysis we included up to four tests over time from each building.

Data sources

We obtained data on the public water service areas compiled by the PADEP from the Pennsylvania State University's Spatial Data Access website (www.pasda.psu.edu). Any home outside the public water supplier's service area was assumed to use well water. Statewide bedrock geology and physiographic sections were downloaded as shapefiles from the Pennsylvania Department of Conservation and Natural Resources (PADCNR) (www.dcnr.state.pa.us). On average, each geologic unit covers 749 non-contiguous km^2 . One important geologic unit is the Felsic gneiss, which is found throughout the state. The Reading Prong section primarily contains Felsic gneiss, however the section is present in only three counties, identified as Reading Prong counties in Figure 2.

We downloaded monthly average temperature and rainfall in ten regions from the Pennsylvania State Climatologist (<http://climate.psu.edu>). Based on 2000 U.S. Census boundary files, buildings were assigned to a minor civil division: cities, moderate to high-density boroughs, and suburban and rural townships. Community socioeconomic deprivation, an indicator of community SES, was derived from 6 z-transformed U.S. Census 2000 variables (Schwartz et al. 2011). Marcellus shale development data came from PADEP and PADCNr, from January 1, 2005 until December 31, 2013, with the latitude and longitude of each well, the date of well drilling, natural gas produced, and number of producing days.

Marcellus shale development metrics

The Marcellus Formation is 1,500 to 2,500m underground and underlies a large section of Pennsylvania from the southwest curling northeast. Only unconventional wells (horizontal wells, hydraulic fracturing) were included (Figures 2, 3A, and 3B,).

Spud date was the day well drilling began and production start date was the day the well first produced natural gas. We estimated a start date of production for each well as:

$$\text{Production start date at well } i = (l_p - k_p)I_p \quad [1]$$

where l_p is the last day of production in period p , k_p is the number of days in production in period p , and I_p equals 1 when period p is the first period of production for well i , and 0 otherwise. We estimated daily natural gas production for each well in its first production period as the volume of gas produced in its first period divided by the number of days of reported production. In subsequent periods we estimated daily gas production as the volume of gas reported in each period, divided by the number of days in that production period. When wells were missing one or more production volumes by period, we imputed missing volumes for periods in which there was

data before and after ($n = 102$ wells) assuming a linear decline over time. We imputed missing spud dates ($n = 149$ wells) using conditional mean imputation based on production start date, stimulation (hydraulic fracturing) date, year and geographic region.

Two primary Marcellus development metrics were created based on all wells in the state, one on drilled wells and the other on wells in production. Wells drilled prior to the start of an indoor radon test were included in that building's exposure assignment. Once a well was drilled it was assumed to contribute until the end of the study period, December 31, 2013. We calculated drilled well exposure assignment:

$$\text{Building } j \text{ metric} = \sum_{i=1}^n \sum_{k=1}^l (I_A(k,m)/d_{ij}^2)/m, \quad [2]$$

where n is the number of drilled wells, m is the duration of the indoor radon test in days, k is the day with 1 equal to January 1, 2005 and l is equal to 3287 (to December 31, 2013), $I_A(k,m)$ is 1 when well i has been drilled before day k and the indoor radon test at building j is conducted from day k to day $k + m$, and 0 otherwise, and d_{ij}^2 is the squared-distance between the coordinates of the wellhead of well i and building j . We calculated the producing well exposure assignment:

$$\text{Building } j \text{ metric} = \sum_{i=1}^n \sum_{k=1}^l (I_A(k,m)g_p/d_{ij}^2)/m \quad [3]$$

where n is the number of producing wells, m is the duration of the indoor radon test in days, k is the day with 1 equal to January 1, 2005 and l is equal to 3287 (to December 31, 2013), $I_A(k,m)$ is 1 when well i is producing on day k and the indoor radon test at building j is conducted from day k to day $k + m$, and 0 otherwise, g_p is the estimated amount of natural gas produced (in thousands of m^3) by well i on day k , and d_{ij}^2 is the squared-distance between the coordinates of the wellhead of well i and building j .

Statistical analysis

The goal of the analysis was to evaluate associations of year, county category, geologic unit, community type, community SES, well water use, and metrics of unconventional natural gas development with indoor radon concentrations. Building was the unit of analysis. The distribution of radon concentrations was skewed, so we used \ln -radon concentration as our outcome variable to improve compliance with assumptions of linear regression (i.e., homoscedasticity and normality of residuals). We used one-way ANOVA to assess unadjusted differences in indoor radon concentrations by other covariates. To evaluate associations with indoor \ln -radon concentration we used multivariable linear regression and generalized estimating equations to account for within-building correlation when models included more than one measurement per building. When beta coefficients are <0.1 , $100 \times \beta$ can be interpreted as approximating the percent change in radon concentration associated with a 1-unit change in the independent variable. In models used to assess the spatial distribution of radon levels we wanted to remove the contribution of building-related factors. Models used to assess associations of unconventional natural gas development with radon levels did not contain county, minor civil division, or community SES because of concern about over-adjustment. Covariates were included in models 1-4 because of *a priori* hypotheses that they could confound the relationship between our primary variables of interest and \ln -radon concentration or based on the quasi-likelihood information criterion (Hardin and Hilbe 2013).

Model 1A included only measurements taken on the first test date at each building ($n = 762,725$ buildings and $n = 866,735$ radon values), which included some averaged values when multiple tests were performed on the same floor on the same day. Model 1A was adjusted for test year (1989-2013), test location (basement or first floor), well water use (yes or no), 13 building types

(including “unknown”), test type (listed above), test duration, season, weather (average temperature and rainfall for 10 regions during month radon measurement began with linear, quadratic and cubic terms to account for nonlinearity), minor civil division, county (n = 67), and 179 mutually exclusive geologic units (reference group = Stockton Formation [n = 62,026] plus 12 geologic units with less than 20 tests). We used model 1B to evaluate changes over time in within-building basement radon levels, by estimating model 1A, restricted to basement measurements, from up to 4 testing dates per building (n = 714,097 buildings and n = 1,015,764 radon values). We also assessed changes in radon levels over time for buildings with high initial concentrations by restricting model 1B to buildings with initial radon concentrations ≥ 740 Bq/m³ (n = 55,161 buildings and n = 99,293 radon values).

In model 2, we assessed differential changes in basement radon concentration by place and time by removing county from model 1A and restricting to basement radon values (n = 705,798 buildings and radon values). We ran 5 separate regressions by county category (Philadelphia, Reading Prong [which have no Marcellus activity], low Marcellus activity [<100 wells drilled by 2014], high Marcellus activity [≥ 100 wells drilled by 2014], and no Marcellus activity (Supplementary Material, Table S1). We then plotted the predicted values of the geometric mean radon concentration by county category and year; 95% confidence intervals were estimated using the delta method (Cox 1998).

We produced two maps of statewide basement radon concentrations from 2006-2013. The first displayed median radon concentrations per geologic unit (with ≥ 10 measurements). In the second we removed variability due to building-level factors (which could help target remediation efforts to certain locations). We did this with model 3 by regressing \ln -radon on building-level factors (i.e., year, building type, test type, test duration, season, average temperature and rainfall). In

model 4, we fit a linear regression of the residuals from model 3 on only geologic unit, county, and well water use (n = 304,278 buildings and radon values) and then used model 4 to output new predicted radon concentrations in a 500m x 500m grid statewide. Split samples suggested model 4 predicted well; and residual semivariogram plots did not exhibit spatial autocorrelation.

We used models 5 and 6 to evaluate two *a priori* hypotheses of the possible contribution of unconventional natural gas development on indoor radon concentrations: (1) ambient air could contribute to indoor radon concentrations through the release of radon and radium in the drilling process, primarily in the summer when buildings are more likely to be open; and (2) produced natural gas containing radon could enter building air through use of natural gas for cooking or unvented heating and, given a transit speed of about 16 km/hour in pipelines (Gogolak 1980), all buildings in the state could be affected.

In model 5, we evaluated the associations of the drilled well metric (Equation 2) with \ln -radon concentration by restricting model 1A to: the years 2005-2013 (primary years of Marcellus development); measurements taken only during July, August, and September; and buildings located within 20km of a drilled well at the time of the radon test. Since summer months had little variability in temperature, we did not include temperature in model 5. We also fit model 5 separately for first floor (n = 1044 buildings and radon values) and basement (n = 18,123 buildings and radon values) because of hypotheses about pathways of radon entry. Model 5 excluded 3 first floor and 130 basement radon concentrations from buildings located within 800 meters of a well because we did not have enough data to fit a curve for distances <800m and 9 first floor radon values that were outliers (studentized residuals >3). As a counterfactual analysis, we re-ran model 5 including buildings from 1989-2005 that would be located within 20km of a Marcellus well by December 2013.

To evaluate associations of the producing well metric (Equation 3) and \ln -radon concentrations, in model 6, we restricted model 1A to the years 2005-2013 and excluded buildings located within 800m of a producing well ($n = 315$). Because year and the production well metric were highly correlated ($\rho = 0.95$) the regression models could not separate their independent influence, so we presented model 6 production associations unadjusted and adjusted for year, as well as year associations unadjusted for production. Regression analysis was performed using Stata 13 (StataCorp). We tested for linear trend by year by including year as a continuous variable. Alpha was set at 95% and statistical significance was $p < 0.05$. Exposure metric creation and radon predictions were performed using R version 3.0.0 (R Foundation for Statistical Computing) and the sp package.

Results

Our primary analysis included 866,735 first indoor radon values from 762,725 buildings from 1989-2013. Every county reported results (Table S2) with a median of 3447, ranging from 59 in Forest to 99,590 in Allegheny. The majority (81.4%) of values were from basements ($n = 705,798$) with a median concentration of 118.4 Bq/m³ (IQR = 59.2-262.7); 42.2% of these values in basements ($n = 297,614$) met or exceeded the EPA action level (Table 1). Radon concentration varied within and between county categories across the study period, with Reading Prong counties having significantly higher and Philadelphia significantly lower radon concentrations.

In total, 7469 unconventional natural gas wells were drilled in 39 Pennsylvania counties between 2005-2013 (Figure 3A). Over 5000 of those wells entered production, producing 191 billion cubic meters of natural gas between 2009-2013 (Figure 3B). We identified 1056 buildings with radon values from the first floor, during the summer and located within 20km of a drilled well at the time of test. The median of the drilled well metric of these buildings was 0.6 wells/km² (IQR

= 0.2-1.3). The median of the producing well metric of buildings statewide was 294 m³/day/km² (IQR = 3-4464). There were increasing median radon concentrations across quartiles of the production well metric for both first floor and basement (Table 1).

In unadjusted analysis several variables were associated with indoor radon concentrations: well water, building type, duration of test, season, weather during the test, community SES, community type and county; geologic unit associations were strong with large variation by unit (e.g., Axemann Formation, median = 365 Bq/m³, IQR = 167-679 vs. Stockton Formation, median = 93 Bq/m³, IQR = 52-178, Table 1). Communities with lower SES had lower radon levels, but this variable was not included in subsequent models because of concerns regarding mediation (i.e., drilling improves individual SES and community SES, but richer individuals have more tightly sealed homes and higher radon concentrations).

In adjusted analysis (model 1A, n = 866,735 first basement and first floor values), many variables were associated with radon concentrations. Strong associations were observed for specific geologies, for example Axemann, Bellefonte, and Nittany Formations were associated with 220-250% higher radon concentrations, compared to the Stockton Formation (Table S2). Alpha track (generally long-term) and charcoal liquid scintillation tests were associated with 23% and 27% higher radon levels, respectively, compared to activated charcoal tests. Buildings using well water (vs. municipal water) also had 21% higher concentrations ($\beta = 0.191$, 95% CI: 0.184, 0.198). Buildings in cities vs. townships were associated with lower radon levels ($\beta = -0.323$, 95% CI: -0.333, -0.314). There were non-linear associations of rainfall and temperature; less rainfall and cooler temperatures were generally associated with higher radon concentrations. When up to four temporally-ordered basement measurements per building were evaluated (model 1B, n = 1,015,764), we observed a significant decrease in radon concentration across

tests with a 37.1% (95% CI: 36.7, 37.3) decline from test 1 to 2, 51.5% (95% CI: 51.1, 51.9) from 1 to 3 and 58.0% (95% CI: 57.4, 58.9) from 1 to 4 (Table S3). Among buildings with an initial basement radon concentration ≥ 740 Bq/m³ we observed from the first test an 88.8% (95% CI: 88.6, 88.9) decline to the second test, and a 92.3% (95% CI: 92.1 to 92.4) decline to the third (Table S4).

After controlling for confounding variables including geologic unit (model 2, basement values only), there was evidence of an upward trend from 2004-2012 ($p < 0.001$). Confidence intervals overlapped among the high, low and no Marcellus activity counties, particularly between no activity and high activity counties, before 2004, whereas there was little or no overlap after that time, with high activity counties having the highest estimated radon concentrations, followed by no activity and low activity counties, respectively. However, fewer measurements were taken in earlier years, resulting in less precise estimates with more variation from year to year (Figure 4). It should be noted that when both basement and first floor values were included (model 1A, Table S2) the upward trend began in 2006 ($p < 0.001$). There were large differences across the state in median radon concentrations by geologic unit (Figure 5A). Geologic unit and well water use did not appear to make large contributions to indoor radon concentrations in regions with many drilled Marcellus wells (Figure 5B, models 3-4).

The drilled well metric was significantly associated with first floor summer radon concentrations in buildings located within 20km of a drilled well; for each additional drilled well per km² surrounding the building, first floor radon levels were estimated to be 2.8% higher (drilled well $\beta = 0.028$, 95% CI: 0.001, 0.05) (model 5). We also found a positive, but attenuated, association with basement measurements (drilled well $\beta = 0.010$, 95% CI: 0.003, 0.020). In a sensitivity analysis, there was no association between the counterfactual drilled well metric for future wells

and summer first floor concentrations between 1989-2005 (drilled well $\beta = 0.001$, 95% CI: -0.022, 0.024).

The producing well metric was not associated with indoor radon concentration when year was included in model 6 (production $\beta = -0.001$, 95% CI: -0.003, 0.002); when year was not, gas production was significantly associated with indoor radon concentration and radon concentrations were estimated to be 1.3% higher with each additional 100 m³ of natural gas produced per day per km² (production $\beta = 0.013$, 95% CI: 0.005, 0.020). There was a positive association between year and radon concentrations between 2005-2013 when the production metric was removed from model 6 (year $\beta = 0.012$, 95% CI: 0.011, 0.014).

Discussion

We identified several predictors of indoor radon concentrations in Pennsylvania, a state with historically high levels (Alter and Oswald 1987). Water source, building type, test type, test duration, season, weather, county, and geologic unit were associated with indoor radon concentration. When data were aggregated to county categories, on average, Reading Prong counties had the highest indoor radon concentrations. Nearly 300,000 homes had a first basement test result that exceeded the EPA action level. We observed fluctuating radon concentrations throughout the study period; low Marcellus activity counties consistently had lower radon concentrations than both high and no Marcellus activity counties, before and after drilling began. Then from 2005-2013 the high activity counties had higher basement radon levels than both low and no Marcellus activity counties with confidence intervals that did not overlap, and there was evidence of a significant upward trend. In a model using first floor and basement values and adjusting for each county (model 1A), radon concentrations only began increasing in 2006. When we included multiple basement measurements per building, radon levels declined with

repeated measurements within a building, which is good news for public health and suggests state remediation programs are effective.

Buildings located in cities had nearly 27% lower radon levels than those located in more rural townships. Previous work suggests this difference is not due to weatherization of homes (Cohen and Gromicko 1988); it may occur because cities are sited in low-lying, alluvial sites where radon levels are low (Briggs et al. 2008). However, the association persisted after adjustment for geologic unit and community SES. Buildings located in poorer communities also tended to have lower radon concentrations, consistent with past research (Cohen and Gromicko 1988).

We found that buildings using well water had 21% higher indoor radon concentrations than those using municipal water. The release of waterborne radon during showering or washing can contribute to concentrations in buildings. The National Research Council has estimated that 10,000 pCi/L (37,000 Bq/m³) of waterborne radon entering a building is needed to increase indoor air concentration by 1 pCi/L (37 Bq/m³) (NRC 1999). Our 20% increase represented approximately 37 Bq/m³. An early study of Pennsylvania groundwater wells reported only 10% exceeded 185,000 Bq/m³ (Swistock et al. 1993), putting our estimate at odds with the rule-of-thumb.

We found a statistically significant association between proximity to unconventional natural gas wells drilled in the Marcellus shale and first floor radon concentration in the summer, with a positive, but attenuated association for basement levels, which suggests a pathway through outdoor ambient air, but does not rule out the possibility of radon moving from the basement to the first floor. Geographic location did not appear to account for the association since we did not find an association in buildings prior to 2006 that would be located near Marcellus wells in the

future. Prior studies suggest that levels of radon in ambient air are low; our finding merits further study because the Marcellus shale is known to contain elevated levels of uranium (EPA 2008), and flowback water and reserve pit soil can contain elevated levels of radium, which could create an environmental exposure pathway (Rich and Crosby 2013; Rowan et al. 2011; Warner et al. 2012). It is also possible that radon could enter buildings through the use of natural gas containing radon. However, concentrations at the wellhead in Pennsylvania have a median of 1369 Bq/m³ (Rowan and Kraemer 2012), much lower than the 37,000 Bq/m³ thought needed to increase radon concentrations by 12.2 Bq/m³ annually in homes that use gas appliances (Gogolak 1980). Our findings should be interpreted in the context of Pennsylvania's recent "Technologically Enhanced Naturally Occurring Radioactive Material (TENORM)" study report from January 2015, which concluded: "There is little potential for additional radon exposure to the public due to the use of natural gas extracted from geologic formations in Pennsylvania" (Perma-Fix Environmental Services Inc., 2015). However, the study did detect radon in several components of the UNGD process and waste stream, such as natural gas, drill cuttings, and wastewater.

Our analysis had several limitations. We had no information on radon-resistant construction, construction year, types of remediation completed, type of heating and cooking systems, quantity of natural gas and water used in the building, degree of sealing of the building for energy efficiency, soil type near the building, wind speed and direction, and individual SES. This missing data makes attributing increased radon levels to a particular source difficult. For instance, it is possible that the observed upward trend from 2004-2012 was simply the result of buildings being sealed more tightly during this time.

We did not know whether a radon professional or a homeowner performed each radon test. However, homes are usually tested during real estate transactions, and radon professionals generally perform these tests ensuring impartial results. Tests are also performed when people are worried about their levels or want to retest after abatement. Worry about levels could introduce a form of selection bias sometimes observed in universal screening programs in which those with higher radon levels would be more likely to test first, which would account for the temporal trends up to 2005. We addressed the abatement concern by only including first measurements. Additionally, this analysis should be considered exploratory since we did not perform any environmental radon measurements, specifically directed at evaluating Marcellus or well water hypotheses.

Conclusion

Radon continues to be a concern in Pennsylvania, and geology is an important contributor. Well water may contribute more to indoor radon than previously thought. There has also been a general rise in concentrations since 2006. The measurements of the Pennsylvania TENORM study should be periodically repeated given the projection of 60,000 wells in Pennsylvania by 2030 (Johnson et al. 2010). Future studies of building radon levels include more information about buildings, including age, heating systems, remediation intervention, and radon-resistant construction. Radon exposure represents a major environmental health risk, and in addition to future studies to understand the impact of drilling on radon levels, there is continuing need for a radon program in Pennsylvania to track and evaluate radon concentrations and to encourage testing and remediation.

References

- Alter HW, Oswald RA. 1987. Nationwide distribution of indoor radon measurements: a preliminary data base. *JAPCA* 37(3):227-231.
- Briggs D, Abellan JJ, Fecht D. 2008. Environmental inequity in England: small area associations between socio-economic status and environmental pollution. *Soc Sci Med* 67(10):1612-1629.
- Cohen BL, Gromicko N. 1988. Variation of radon levels in U.S. homes with various forms. *JAPCA* 38(2):129-134.
- Cox C. 1998. Delta method. In: *Encyclopedia of Biostatistics*, (Armitage P, Colton T, eds). New York:John Wiley, 1125 - 1127.
- Darby S, Hill D, Auvinen A, Barros-Dios JM, Baysson H, Bochicchio F, et al. 2005. Radon in homes and risk of lung cancer: collaborative analysis of individual data from 13 European case-control studies. *BMJ* 330(7485):223.
- EPA (Environmental Protection Agency). 1992. Technical support document for the 1992 citizen's guide to radon; EPA-400r-92-11. Washington, D.C. Available: <http://nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=000001L3.TXT> [accessed 8 Mar 2015].
- EPA (Environmental Protection Agency). 2008. Technologically Enhanced Naturally Occurring Radioactive Materials From Uranium Mining; EPA 402-R-08-005. Washington, DC. Available: <http://www.epa.gov/radiation/docs/tenorm/402-r-08-005-voli/402-r-08-005-v1.pdf> [accessed 8 Mar 2015].
- Folger PF, Nyberg P, Wanty RB, Poeter E. 1994. Relationships between Rn-222 dissolved in ground water supplies and indoor Rn-222 concentrations in some Colorado front range houses. *Health Phys* 67(3):245-253.
- Gogolak CV. 1980. Review of 222-Rn in Natural Gas Produced from Unconventional Sources. New York, NY:Department of Energy. Available: <http://treichlerlawoffice.com/radiation/DOEuranium653239.pdf> [accessed 8 Mar 2015].
- Gross EL. 2013. Distribution of indoor radon concentrations in Pennsylvania, 1990-2007: U.S. Geological Survey Scientific Investigations Report 2013–5143. Available: <http://pubs.usgs.gov/sir/2013/5143/> [accessed 1 June 2014].

- Gundersen LC. 1991. Radon in sheared metamorphic and igneous rocks. In: Field Studies of Radon in Rocks, Soils and Water, (Gundersen LC, Wanty RB, eds). United States CRC Press, 39-49.
- Hardin JW, Hilbe JM. 2013. Generalized Estimating Equations. Second ed. Boca Raton, FL: CRC Press.
- Johnson Jr R, Bernhardt D, Nelson NS, Calley Jr H. 1973. Assessment of potential radiological health effects from radon in natural gas. Environmental Protection Agency, Washington, DC (USA). Office of Radiation Programs. Available: nepis.epa.gov/Exe/ZyPURL.cgi?Dockey=91012JNH.TXT [accessed 8 Mar 2015].
- Johnson N, Gagnolet T, Ralls R, Zimmerman E, Eichelberger B, Tracey C, et al. 2010. Pennsylvania Energy Impacts Assessment, Report 1: Marcellus Shale Natural Gas and Wind. Available: http://www.nature.org/media/pa/pa_energy_assessment_report.pdf [accessed 2 July 2014].
- NAS (National Academy of Sciences). 1999. Health Effects of Exposure to Radon: Biological Effects of Ionizing Radiation (BEIR) VI Report. Washington, DC: National Academy Press.
- NRC (National Research Council). 1999. Risk Assessment of Radon in Drinking Water. Washington, DC: The National Academies Press.
- Pawel DJ, Puskin JS. 2004. The U.S. Environmental Protection Agency's assessment of risks from indoor radon. *Health Phys* 87(1):68-74.
- Perma-Fix Environmental Services Inc. for Pennsylvania Department of Environmental Protection. 2015. Technologically Enhanced Naturally Occuring Radioactive Materials (TENORM) Study Report. Beaver, Pennsylvania. Available: http://www.elibrary.dep.state.pa.us/dsweb/Get/Document-105822/PA-DEP-TENORM-Study_Report_Rev._0_01-15-2015.pdf [accessed 12 Mar 2015].
- Rich AL, Crosby EC. 2013. Analysis of reserve pit sludge from unconventional natural gas hydraulic fracturing and drilling operations for the presence of technologically enhanced naturally occurring radioactive material (TENORM). *New Solut* 23(1):117-135.
- Rowan EL, Kirby C, Engle E, Kraemer T. 2011. Radium Content of Oil- and Gas-Field Produced Waters in the Northern Appalachian Basin (USA) - Summary and discussion of data: U.S. Geological Survey Scientific Investigations Report 2011-5135. Available: <http://pubs.usgs.gov/sir/2011/5135/> [accessed 25 June 2014].

- Rowan EL, Kraemer T. 2012. Radon-222 Content of Natural Gas Samples from Upper and Middle Devonian Sandstone and Shale Reservoirs in Pennsylvania: Preliminary Data. Available: <http://pubs.usgs.gov/of/2012/1159> [accessed 30 June 2014].
- Schwartz BS, Stewart WF, Godby S, Pollak J, DeWalle J, Larson S, et al. 2011. Body mass index and the built and social environments in children and adolescents using electronic health records. *Am J Prev Med* 41(4):e17-e28.
- Swistock BR, Sharpe WE, Robillard PD. 1993. A survey of lead, nitrate and radon contamination of private individual water-systems in Pennsylvania. *J Environ Health* 55(5):6-12.
- UNSCEAR (United Nations Scientific Committee on the Effects of Atomic Radiation). 2009. UNSCEAR 2006 Report, Annex E. Sources-to-Effects Assessment for Radon in Homes and Workplaces. United Nations, New York. Available: http://www.unscear.org/docs/reports/2006/09-81160_Report_Annex_E_2006_Web.pdf [accessed 8 Mar 2015].
- Warner NR, Jackson RB, Darrah TH, Osborn SG, Down A, Zhao K, et al. 2012. Geochemical evidence for possible natural migration of Marcellus Formation brine to shallow aquifers in Pennsylvania. *Proc Natl Acad Sci U S A* 109(30):11961-11966.
- WHO (World Health Organization). 2009. Handbook on Indoor Radon: A Public Health Perspective. 9241547677. Geneva, Switzerland:WHO Press. Available: http://www.nrsb.org/pdf/WHO_Radon_Handbook.pdf [accessed 8 Mar 2015].

Table 1. Radon concentrations by test and building characteristics, stratified by test location.

Variable	Category	Basement*			First floor*			
		n (%)	Median (IQR), Bq/m ³	Range, Bq/m ³	n (%)	Median (IQR), Bq/m ³	Range, Bq/m ³	
All results	Total	705798 (100)	118.4 (59.2-262.7)	0-69057	160937 (100)	55.5 (29.6-111.0)	0-111481	
EPA action level	<148 Bq/m ³	408184 (57.8)	66.6 (40.7-99.9)	0-147.9	131245 (81.6)	44.4 (25.9-74)	0-147.9	
	≥148 Bq/m ³	297614 (42.2)	310.8 (207.2-580.9)	148-69057	29692 (18.5)	262.7 (188.7-447.7)	148-111481	
Well water use	No	591565 (83.8)	111.0 (58.1-236.8)	0-69057	138804 (86.3)	51.8 (27.8-103.6)	0-111481	
	Yes	114233 (16.2)	185.0 (81.4-458.8)	0-55463	22133 (13.8)	74.0 (37.0-164.7)	0-14822	
Building type	2-Story	298672 (42.3)	114.7 (61.1-114.7)	0-55463	73340 (45.6)	53.65 (29.6-103.6)	0-111481	
	3-Story	69008 (9.8)	166.5 (77.6-166.5)	2.2-33973	8837 (5.5)	70.3 (33.3-162.8)	0.7-7478	
	Apartment	1999 (0.3)	82.0 (44.4-173.9)	3.7-5254	1042 (0.7)	33.3 (18.5-68.5)	0.7-1395	
	Bi-level	12599 (1.8)	131.3 (62.9-294.2)	1.1-25937	2628 (1.6)	77.7 (37.0-166.5)	1.9-9476	
	Cape Cod	15801 (2.2)	127.7 (70.3-257.2)	0-29637	3837 (2.4)	59.2 (29.6-103.6)	0-29711	
	Commercial	1773 (0.3)	77.7 (42.6-157.3)	3.7-4449	871 (0.5)	40.7 (22.2-83.9)	0.7-6915	
	Contemporary	4156 (0.6)	136.0 (66.6-296)	3.7-25530	1968 (1.2)	51.8 (25.9-108.8)	3.7-2760	
	Public/School	370 (0.1)	94.7 (47.2-203.5)	13.6-5176	202 (0.1)	51.8 (27.1-96.2)	3.7-636	
	Ranch	63946 (9.1)	151.7(79.6-323.2)	0.9-69057	14764 (9.2)	66.6 (37.0-136.9)	0-10286	
	Split level	17788 (2.5)	107.3 (59.2-218.3)	1.5-41607	5822 (3.6)	59.2 (33.3-107.3)	0-8251	
	Townhouse	42691 (6.1)	68.5 (40.7-125.8)	0.2-32751	16920 (10.5)	37.0 (22.2-66.6)	0-22459	
	Trailer	183 (0.03)	88.8 (51.8-192.4)	18.5-2531	139 (0.1)	33.3 (18.5-33.3)	3.7-662	
	Unknown	176812 (25.1)	122.1 (55.5-297.9)	0-35668	30567 (19.0)	62.9 (29.6-153.0)	0-16119	
	Test type	Activated charcoal	237932 (33.7)	129.5 (55.5-325.6)	0-69057	54957 (34.2)	59.2 (25.9-142.5)	0-50294
		Alpha track	7074 (1.0)	161.1 (81.4-333.0)	0.7-14796	1844 (1.2)	99.9 (42.7-221.4)	0.4-3441
Charcoal liquid scintillation		44936 (6.4)	162.8 (70.3-392.2)	0-32751	4934 (3.1)	77.7 (33.3-186.9)	3.7-16119	
Continuous		209994 (29.8)	114.7 (59.2-236.8)	0.2-41544	14647 (9.1)	48.1 (25.9-92.5)	0.1-111481	
Electret ion chamber		205862 (29.2)	111.0 (62.9-214.6)	0-62974	84555 (52.5)	53.65 (29.6-99.9)	0-29711	
Test duration	1-7 days	693864 (98.3)	118.4 (59.2-262.7)	0-69057	157912 (98.2)	55.5 (29.6-111.0)	0-111481	
	≥8 days	11934 (1.7)	148.0 (74.0-310.8)	0-69057	3025 (1.8)	81.4 (37.0-181.3)	0-3593	
Season	Winter	169921 (24.1)	114.7 (59.2-247.9)	0-55463	37886 (23.5)	48.1 (25.9-96.2)	0-50294	
	Spring	198485 (28.1)	114.7 (59.2-229.4)	0-62974	46432 (28.9)	51.8 (27.8-98.1)	0-22496	
	Summer	174007 (24.7)	133.2 (66.6-299.7)	0-41543.6	40320 (25.1)	66.6 (33.3-136.9)	0-111481	
	Autumn	163385 (23.2)	118.4 (59.2-292.3)	0-69057	36886 (22.6)	59.2 (29.2-129.5)	0-29711	
Average temperature in month of test (°C)	<0	84259 (11.9)	3.3 (1.6-8.0)	0.004-930	17294 (10.8)	1.6 (0.8-3.7)	0-276	
	0 to <10	232372 (32.9)	3.3 (1.6-7.7)	0-1866	53651 (33.3)	1.6 (0.8-3.4)	0-3013	
	10 to <18.3	189693 (26.9)	3.4 (1.7-7.3)	0-1499	43018 (26.7)	1.5 (0.8-3.0)	0-607	
	≥18.3	199474 (28.3)	3.0 (1.6-6.2)	0-1702	46974 (29.2)	1.4 (0.7-2.6)	0-608	

Variable	Category	Basement*			First floor*		
		n (%)	Median (IQR), Bq/m ³	Range, Bq/m ³	n (%)	Median (IQR), Bq/m ³	Range, Bq/m ³
Average rainfall in month of test (cm)	<7.1	236239	125.8 (62.9-281.9)	0-69056	53693	59.2 (29.6-121.0)	0-50294
	7.2-10.8	232866	116.6 (59.2-251.6)	0-62974	55928	55.5 (29.6-107.3)	0-111481
	≥10.9	236693	116.6 (59.2-255.3)	0-41607	51316	53.7 (27.8-107.3)	0-22496
Community socioeconomic deprivation quartile ^a	1 (<-4.9)	169327 (24.5)	118.4 (59.2-262.7)	0-35897	46100 (29.4)	59.2 (29.6-118.4)	0-50294
	2 (-4.9 to -3.3)	172068 (24.9)	133.2 (66.6-306.0)	0-55463	37389 (23.8)	62.9 (33.3-129.5)	0-29711
	3 (-3.2 to -1.1)	177619 (25.7)	129.5 (66.6-284.9)	0-69057	36734 (23.4)	59.2 (29.6-114.7)	0-18537
	4 (≥-1.0)	173407 (25.0)	103.6 (53.7-222.0)	0-35668	36742 (23.4)	44.4 (24.1-92.5)	0-111481
Minor civil division	Township	488168 (69.2)	130.7 (64.8-299.7)	0-55463	116311 (72.3)	59.2 (29.6-122.1)	0-50294
	Borough	133990 (19.0)	112.9 (59.2-233.1)	0-69057	25643 (15.9)	51.8 (25.9-103.6)	0-22496
	City	83638 (11.9)	79.6 (44.4-1480)	0-31361	18983 (11.8)	40.7 (22.2-70.7)	0-111481
County category ^b							
No Marcellus activity	Other counties	379223 (53.7)	120.3 (59.2-273.8)	0-62974	112252 (69.8)	55.5 (29.6-111.0)	0-50294
Low Marcellus activity	<100 drilled wells by 2013	174216 (24.7)	114.7 (62.9-233.1)	0-30621	22734 (14.1)	55.5 (27.4-118.4)	0-22496
High Marcellus activity	≥100 drilled wells by 2013	57814 (8.2)	129.5 (70.3-260.9)	0-30858	5753 (3.6)	62.9 (33.3-129.5)	2.6-111481
Reading Prong	Berks, Lehigh, and Northampton	62635 (8.9)	192.4 (85.1-425.5)	0-69057	9632 (6.0)	96.2 (44.4-210.9)	0-14822
Philadelphia	Philadelphia	31910 (4.5)	62.9 (37.0-105.5)	0-31361	10566 (6.6)	37.0 (22.2-62.9)	0-2331
Drilled well within 20km of building	No	637317 (90.3)	118.4 (59.2-266.4)	0-69057	156731 (97.4)	55.5 (29.6-111.0)	0-50294
	Yes	68481 (9.7)	124.0 (70.3-244.2)	0-38658	4206 (2.6)	59.2 (33.3-120.3)	3.7-111481
Drilled well exposure quartile ^c							
1	<0.19 well/km ²	17086 (25.0)	120.3 (70.3-225.7)	3.7-23465	1086 (25.8)	70.3 (37.0-133.2)	3.7-2742
2	0.19 to 0.61 well/km ²	17099 (25.0)	125.8 (70.3-255.3)	18.5-29637	1073 (25.5)	59.2 (29.6-114.7)	18.5-8251
3	0.62 to 1.4 well/km ²	17126 (25.0)	125.8 (70.3-247.9)	18.5-30858	1046 (24.9)	55.5 (29.6-107.3)	14.8-3559
4	>1.4 well/km ²	17170 (25.1)	125.8 (70.3-247.9)	18.5-19769	1001 (23.8)	59.2 (37.0-122.1)	18.5-111481
Producing well exposure quartile ^d							
1	<2.55 m ³ /day/km ²	83971 (24.3)	111.0 (55.5-247.9)	2.6-40928	13052 (31.2)	51.8 (26.3-99.9)	0-8131
2	2.55 to 294.4 m ³ /day/km ²	86196 (24.9)	120.3 (61.1-266.4)	7.4-35897	10826 (25.9)	59.2 (30.9-118.4)	0-29711
3	294.5 to 4312.6 m ³ /day/km ²	86989 (25.1)	125.8 (62.9-281.2)	11.1-62974	10034 (24.0)	59.2 (33.3-122.1)	3.7-12119
4	>4312.7 m ³ /day/km ²	89143 (25.7)	133.2 (70.3-288.6)	11.1-30858	7879 (18.9)	61.1 (33.3-124.0)	5.6-111481

*Categories of all variables in the row headings had statistically significantly different *ln*-radon concentrations by analysis of variance (ANOVA).

^aNot available for buildings located in Philadelphia or Pittsburgh. Community socioeconomic deprivation was assigned at the township, borough, or census tract level, based on 6 indicators derived from the 2000 U.S. Census A: combined less than high school education, not in the labor force, in poverty, on public assistance, civilian unemployment, and does not own a car; a higher score represents a more deprived community. ^bOther counties: Adams, Bedford, Bucks, Carbon, Chester, Cumberland, Dauphin, Delaware, Erie, Franklin, Fulton, Juniata, Lancaster, Lebanon, Mifflin, Montgomery, Montour, Northumberland, Perry, Pike, Schuylkill, Snyder, Union, and York; Marcellus low exposure counties: Allegany, Beaver, Blair, Cambria, Cameron, Centre, Clarion, Columbia, Crawford, Elk, Forest, Huntingdon, Indiana, Jefferson, Lackawanna, Lawrence, Luzerne, McKean, Mercer, Potter, Somerset, Sullivan, Venango, Warren, and Wayne; Marcellus high exposure counties: Armstrong, Bradford, Butler, Clinton, Clearfield, Fayette, Greene, Lycoming, Susquehanna, Tioga, Washington, Westmoreland, and Wyoming. ^cRestricted to 2005-2013 and buildings within 20km of a drilled well at the time of the radon test. ^dRestricted to 2005-2013.

Figure Legends

Figure 1. Flow diagram of radon tests included in six primary models

Figure 2. County category groupings, the Reading Prong Section, and location of spudded Marcellus wells (through 2013)

Figure 3. A. Cumulative and number of new unconventional wells drilled annually, 2005-2013. B. Unconventional natural gas produced in 100 millions of cubic meters, 2005-2013.

Figure 4. Geometric mean and 95% confidence intervals for indoor basement radon concentrations in five county categories, 1989-2013. High Marcellus shale counties had at least 100, and low Marcellus shale counties had 1-100, unconventional wells drilled by 2013. Predicted values were generated from 5 separate linear regression models (one for each county category) including only measurements taken on the first test date at each building ($n = 705,798$ values), adjusted for test year (1989-2013), well water use, 13 building types, 5 test types, test duration, season, weather (average temperature and rainfall with linear, quadratic and cubic terms), minor civil division, and 179 mutually exclusive geologic units (model 2).

Figure 5. A. Unadjusted median basement radon concentrations ($n = 304,278$ tests) in Pennsylvania by geologic unit, 2006-2013. B. Predicted contribution to basement radon concentration from geologic unit, county, and well water after accounting for variation due to year (2006-2013), building type, test type, test duration, season, average temperature and average rainfall (based on models 3-4, $n = 304,278$ values).

Figure 1.

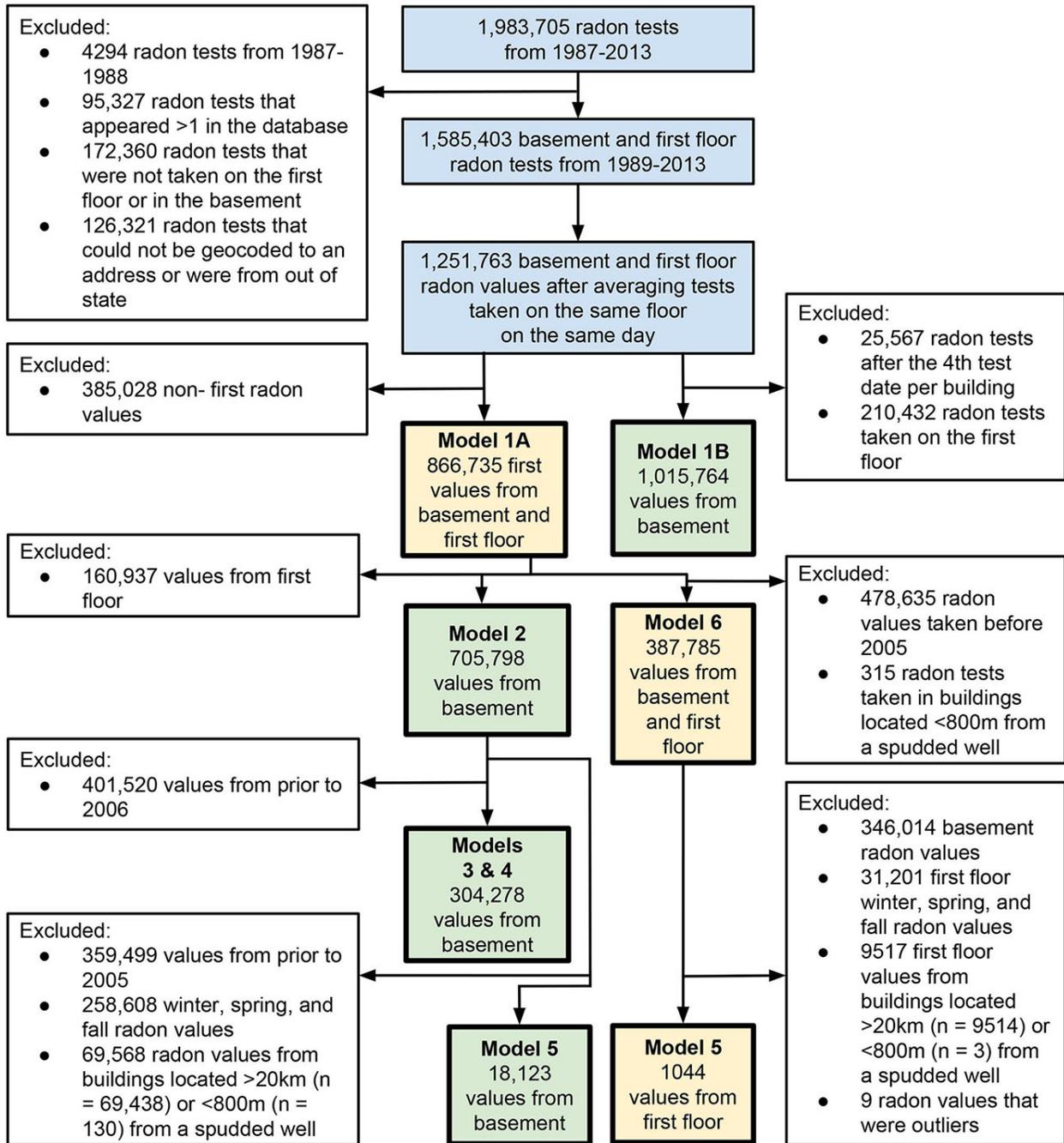


Figure 2.

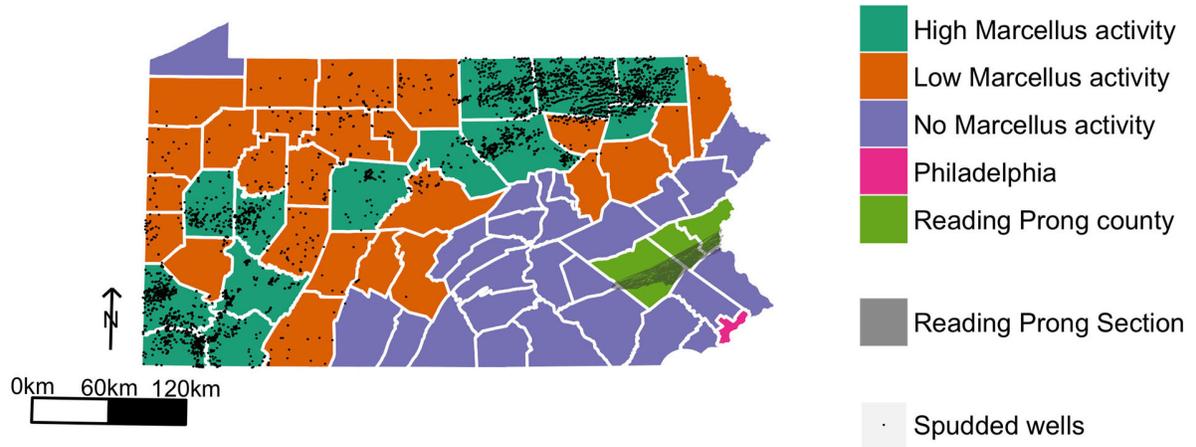


Figure 3.

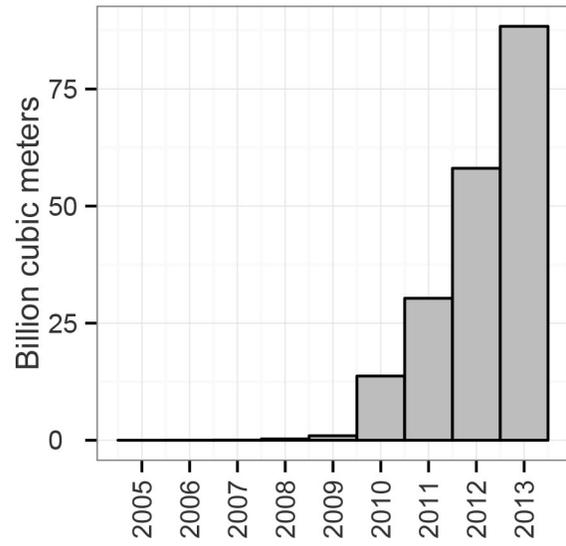
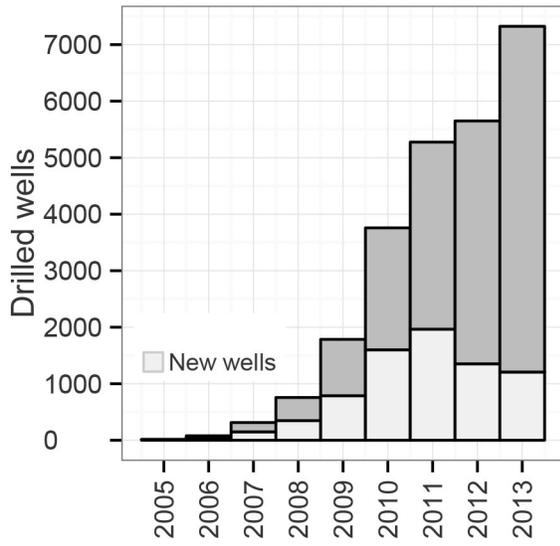


Figure 4.

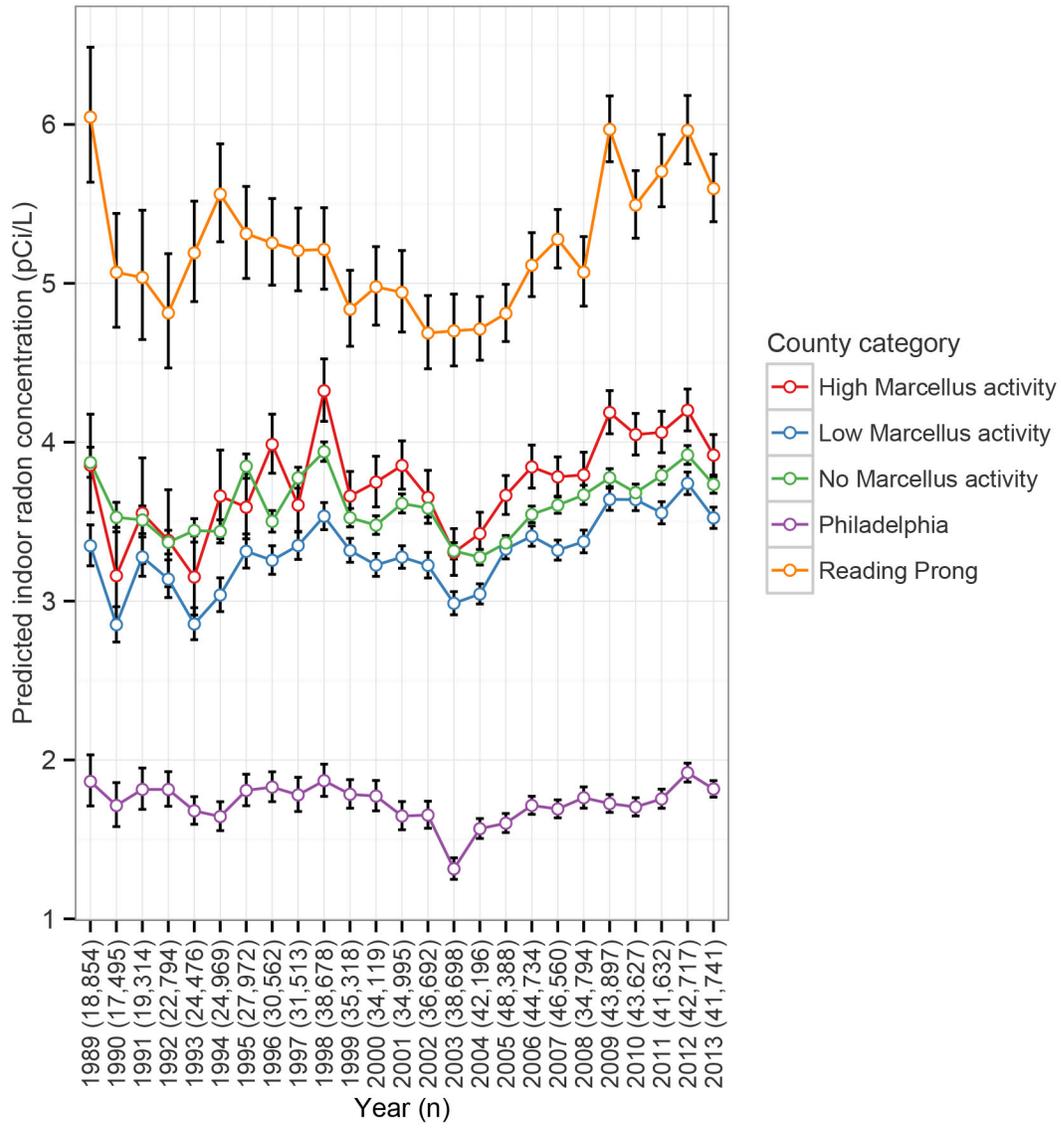
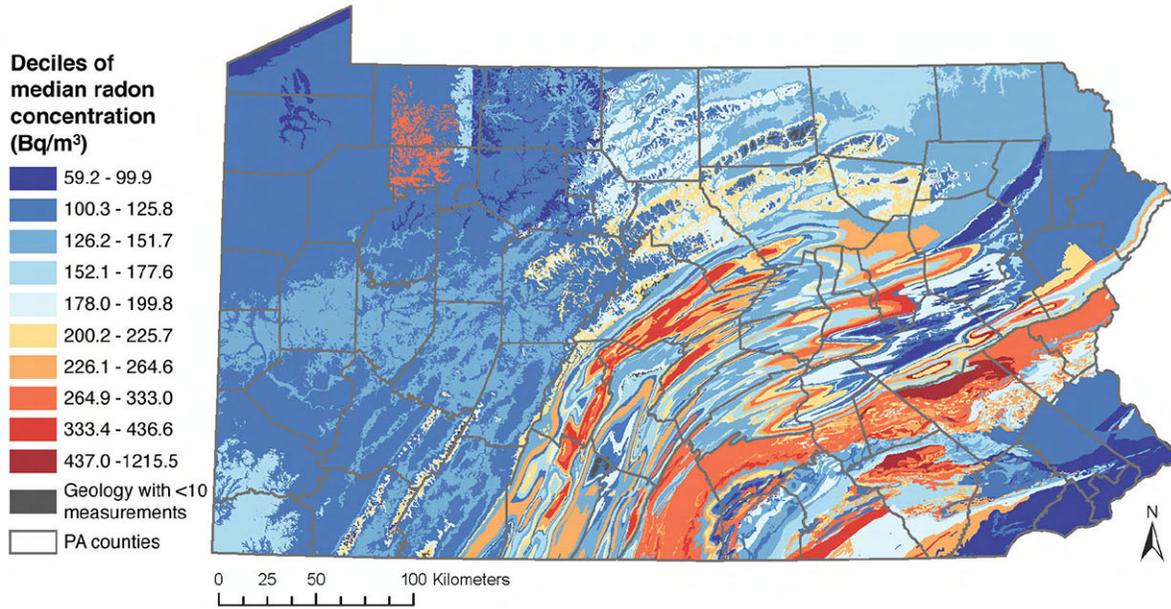


Figure 5.

A.



B.

